

FORM PTO-199 (REV 5-98)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER
				Mo-4805/LeA 31,454
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				097077914
INTERNATIONAL APPLICATION NO.	PCT/EP 96/05335	INTERNATIONAL FILING DATE	12/02/96	To be Assigned
				PRIORITY DATE CLAIMED
				12/13/95
TITLE OF INVENTION PROCESS FOR PREPARING RIGID FORMED MATERIALS CONTAINING URETHANE GROUPS				
APPLICANT(S) FOR DO/EO/US				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.      2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.      3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).      4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.      5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))          a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).          b. <input type="checkbox"/> has been transmitted by the International Bureau.          c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).      6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).      7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(0))          a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).          b. <input type="checkbox"/> have been transmitted by the International Bureau.          c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.          d. <input type="checkbox"/> have not been made and will not be made.      8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).      9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(6)).      10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p>				
Items 11. to 16. below concern other document(s) or information included:				
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.				
12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.				
13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.				
14. <input type="checkbox"/> A substitute specification.				
15. <input type="checkbox"/> A change of power of attorney and/or address letter.				
16. <input type="checkbox"/> Other items or information:				
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<u>Donna J. Veatch</u> (Name of person mailing paper or fee) <u>Donna J. Veatch</u> Signature of person mailing paper or fee)				

17. <input checked="" type="checkbox"/> The following fees are submitted:	INTERNATIONAL APPLICATION NO. PCT/EP 96/05335	ATTORNEY DEPOSITORY 40-4805/LeA 31,454		
Basic National Fee (37 CFR 1.492(a)(1)-(5)); Search Report has been prepared by the EPO or JPO..... \$930.00		<b>CALCULATIONS</b> <small>PTO USE ONLY</small>		
International preliminary examination fee paid to USPTO (37 CFR 1.482) ..... \$720.00				
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).. \$790.00				
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$1,070.00				
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... \$ 98.00				
<b>ENTER APPROPRIATE BASIC FEE AMOUNT</b> =		\$ 930.00		
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$		
<b>CLAIMS</b>		<b>Number Filed</b>	<b>Number Extra</b>	<b>Rate</b>
Total Claims	11	-20 =		X \$22.00
Independent Claims	2	-3 =		X\$82.00
Multiple dependent claim(s) (if applicable)		+270.00		
<b>TOTAL OF ABOVE CALCULATIONS</b>		= \$ 930.00		
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$		
<b>SUBTOTAL</b>		= \$ 930.00		
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(0)).		+ \$		
<b>TOTAL NATIONAL FEE</b>		= \$ 930.00		
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		+ \$ 40.00		
<b>TOTAL FEES ENCLOSED</b>		= \$ 970.00		
		<b>Amount to be: refunded \$ charged \$</b>		
<b>a.</b> <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.				
<b>b.</b> <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>13-3848</u> in the amount of <u>\$ 970.00</u> to cover the above fees. A duplicate copy of this sheet is enclosed.				
<b>c.</b> <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No <u>13-3848</u> . A duplicate copy of this sheet is enclosed.				
<b>NOTE:</b> Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.				
 SIGNATURE				
<u>Lyndanne M. Whalen</u> NAME				
Reg. No. 29,457				
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PATENT APPLICATION  
Mo-4805  
LeA 31,454

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF )  
KARL W. DIETRICH ET AL )  
SERIAL NUMBER: TO BE ASSIGNED )  
FILED: HEREWITH )  
TITLE: PROCESS FOR PREPARING )  
RIGID FOAMED MATERIALS )  
CONTAINING URETHANE )

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231  
Sir:

Upon the granting of a Serial Number for the enclosed application, please amend this application as follows:

IN THE CLAIMS:

Please cancel Claims 1-10 and add the following new Claims 11-21:

-- 11. A process for the production of a rigid polyurethane foamed plastic comprising reacting

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Donna J. Veatch  
(Name of person mailing paper or fee)  
Donna J. Veatch  
Signature of person mailing paper or fee)

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- a) a polyol,
- b) a polyisocyanate,
- c) a blowing agent comprising
  - 1) from about 5 to about 50 parts by weight of a C<sub>3</sub> and/or C<sub>4</sub> alkane and
  - 2) from about 50 to about 95 parts by weight of cyclopentane, and optionally
- d) auxiliary additives.

12. The process of Claim 11 in which c) 1) is n-butane.
13. The process of Claim 11 in which c) 1) is isobutane.
14. The process of Claim 11 in which c) further includes from about 0.5 to about 4 parts by weight of water.
15. The process of Claim 11 in which c) further includes from about 1.5 to about 3 parts by weight of water.
16. The process of Claim 14 in which a) includes from about 5 to about 80 parts by weight of an aromatic amine initiated polyol.
17. The process of Claim 11 in which a) includes from about 5 to about 80 parts by weight of an aromatic amine initiated polyol.
18. The process of Claim 11 in which a) includes from about 20 to about 65 parts by weight of an aromatic amine initiated polyol.
19. A blowing agent composition comprising
  - a) from about 5 to about 50 parts by weight of a C<sub>3</sub> and/or C<sub>4</sub> alkane and
  - b) from about 50 to about 95 parts by weight of cyclopentane.
20. The process of Claim 19 in which a) is n-butane and/or isobutane.
21. A rigid polyurethane foam plastic produced by the process of Claim 11. --

**REMARKS**

Claims 1-10 have been cancelled and rewritten as new Claims 11-21 in an effort to place the claims in better form.

An action on the merits of this application is requested.

Respectfully submitted,

KARL W. DIETRICH  
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CRAPS

- 1 -

A method of producing rigid foamed plastics containing urethane groups

It is known to blow polyurethane rigid foamed plastics, using low-boiling alkanes.

5 It is advantageous to use cyclic alkanes for this purpose since, owing to their low thermal conductivity in the gaseous state, they greatly improve the thermal conductivity of the foamed plastic. The main substance used is cyclopentane. Cyclopentane, however, owing to its relatively high boiling-point (49°C), has the disadvantage that it condenses at low temperatures, which occur normally when

10 polyurethane rigid foamed plastic is used as an insulating material in domestic refrigerators. At these low temperatures, especially when used in refrigerators, cyclopentane does not have its full insulating effect, and also the undesired condensation of the blowing agent results in a negative pressure in the cell, which has to be compensated by increasing the rigidity of the foam or increasing the bulk density.

15

The object of the invention is to provide cyclopentane-containing blowing-agent mixtures which retain the good thermal insulation of cyclopentane even at low temperatures.

20 It has now surprisingly been found that by adding small proportions of low-boiling alkanes in the C<sub>3</sub> and C<sub>4</sub> series, the advantageous thermal conductivity of cyclopentane foam can be retained and also the pressure inside the cells can be considerably increased, especially at low temperatures. "Low temperatures" as usual means temperatures below 10°C, e.g. between -30°C and +5°C.

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DONNA J. WEATCH

Name of person mailing paper or this  
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An important feature of blowing-agent mixtures is that they are liquid at room temperature and the normally gaseous C<sub>3</sub> and C<sub>4</sub> components do not increase the vapour pressure of the total mixture above 1 bar. This has been found particularly in the case of mixtures of cyclopentane and n-butane and/or isobutane, so that no complications result in polyurethane processing. An admixture of low-boiling alkanes appreciably impairs the insulating effect but, at proportions of 10 to 25%

by weight, the impairment is surprisingly small. Cyclopentane usually dissolves easily in most polyols used in rigid polyurethane foam. Low-boiling aliphatic alkanes have much worse solubility, so that C<sub>3</sub> and C<sub>4</sub>-alkanes added to cyclopentane reduce the solubility of the blowing agent. In this case it is advantageous to use 5 polyols started on aromatic amines; these have excellent solubility even in the case of acyclic alkanes.

10 The invention accordingly provides a method of producing rigid foamed plastics containing urethane and optionally containing isocyanurate groups, by reaction of polyols, polyisocyanates, blowing agents and optional foam auxiliary substances, characterised in that the blowing agent is a mixture of 5 to 50 parts by weight of C<sub>3</sub> and/or C<sub>4</sub> alkanes and 50 to 95 parts by weight of cyclopentane.

15 The polyols and polyisocyanates in the method according to the invention can be any per se known starting components.

20 The isocyanate components can e.g. be aromatic polyisocyanates such as described by W. Siefken in Justus Liebigs Annalen der Chemie, 562, pages 75 to 136, e.g. substances having the formula



in which

25 n is 2 to 4, preferably 2, and

Q denotes an aliphatic hydrocarbon radical with 2 to 18, preferably 6 to 10 carbon atoms, or a cycloaliphatic hydrocarbon radical with 4 to 15, preferably 5 to 10 carbon atoms, or an aromatic hydrocarbon radical with 6 to 15, preferably 6 to 13 carbon atoms or an araliphatic hydrocarbon radical with 8 to 15, preferably 8 to 13 carbon atoms, e.g. polyisocyanates such as 30 described in DE-OS 28 32 253, pages 10 to 11.

Particular preference as a rule is given to polyisocyanates which are easily obtainable industrially, e.g. 2,4 and 2,6-tolylene diisocyanate or any mixtures of these isomers ("TDI"), or polyphenyl polymethylene polyisocyanates produced by condensation of aniline and formaldehyde and subsequent phosgenation ("crude MDI") or polyisocyanates containing carbodiimide groups, urethane groups, allophanate groups, isocyanurate groups, urea groups or biuret groups ("modified polyisocyanates"), more particularly modified polyisocyanates derived from 2,4 or 2,6-tolylene diisocyanate or from 4,4'- and/or 2,4'-diphenyl methane diisocyanate.

5      10      The starting components for the polyol component are compounds containing at least two hydrogen atoms capable of reacting with isocyanates and having a molecular weight usually between 62 and 20 000. These include compounds containing amino groups, thiol groups or carboxyl groups and also and preferably compounds containing hydroxyl groups, preferably polyethers, polyesters, polycarbonates, polylactones and polyamides, particularly compounds containing 2 to 8 hydroxyl groups, especially those having a molecular weight of 1 000 to 20 000, e.g. compounds containing at least 2, usually 2 to 8, preferably 2 to 4 hydroxyl groups as known per se in the production of polyurethanes and described e.g. in DE-OS 28 32 253, pages 11 to 18. Mixtures of various compounds of this kind can also be used according to the invention.

15      20      Optionally, known additives and auxiliary substances, such as flame retardants, catalysts and foam stabilizers, are also used.

25      The flame retardants are per se known flame retardants, preferably products liquid at 20°C.

30      The foam stabilizers are preferably polyether siloxanes, especially if soluble in water. These compounds usually have a structure in which a copolymer of ethylene oxide and propylene oxide is combined with a polydimethyl siloxane radical. Foam stabilizers of this kind are described e.g. in US-PS 2 834 748, 2 917 480 and 3 629

32 320 28 32 253

308. The catalysts can be those known per se in polyurethane chemistry, such as tert.-amines and/or organometallic compounds.

Use can also be made of reaction retarders, e.g. acid-reacting substances such as 5 hydrochloric acid or organic acid halides, or cell regulators of per se known kind such as paraffins or fatty alcohols or dimethyl polysiloxanes, or pigments or dyes, or stabilizers against ageing and weathering, plasticizers, fungistatic substances and bacteriostats or fillers such as barium sulphate, kieselguhr, carbon black or whiting.

10 Other examples of surface-active additives and foam stabilizers optionally also used according to the invention, cell regulators, reaction retarders, stabilizers, flame-retarding substances, dyes, fillers, fungistatic substances and bacteriostats and details about use and operation of these auxiliary substances are described in Kunststoff-Handbuch, Volume VII, published by Vieweg and Höchtl, Carl Hanser Verlag, 15 Munich 1966, e.g. on pages 121 to 205.

In the production of foam according to the invention, foaming can also be brought about in closed moulds. The reaction mixture is placed in a mould, which can be made of metal e.g. aluminium or plastic e.g. epoxy resin. In the mould, the 20 expandable reaction mixture foams and forms the moulded member. The foaming process in the mould can be managed so that the moulded part has a cellular surface structure. Alternatively it can be managed so that the moulded part has a compact skin and a cellular core. The procedure according to the invention in the first case is to introduce expandable reaction mixture into the mould in an amount sufficient 25 for the resulting foamed plastic just to fill the mould. In the latter case, the amount of reaction mixture introduced into the mould is more than enough to fill the interior of the mould with foamed plastic. In the latter case, operation is by "over-charging". A method of this kind is known e.g. from US-PS 3 178 490 and 3 182 104.

30 The C<sub>3</sub> and/or C<sub>4</sub> alkanes used according to the invention are preferably n-butane and/or isobutane.

0.5 to 4 parts by weight of water can advantageously be used as a co-blowing agent in addition to the alkane mixture according to the invention. It is preferable to use 1.5 to 3 parts by weight of water as the co-blowing agent.

5 Preferably, in the production of rigid polyurethane foamed plastics according to the invention, the polyol component is a mixture of 5 to 80 parts by weight of polyol based on aromatic amines. It is particularly preferable to use 20 to 65 parts by weight of polyol based on aromatic amines.

10 The invention further provides blowing-agent mixtures containing 5 to 50 parts by weight of C<sub>3</sub> and/or C<sub>4</sub> alkanes, preferably n-butane and/or isobutane, and 50 to 95 parts by weight of cyclopentane.

15 The invention also provides use of the rigid foamed plastics produced according to the invention as an intermediate layer in composite components and for foam filling of cavities in domestic refrigerator construction.

20 Preferably the method according to the invention is used for foam filling of cavities in refrigerators and freezers.

25 Alternatively, of course, foamed plastics can be produced by slabstock production or by the per se known double conveyor-belt process.

The rigid foamed plastics obtainable according to the invention are used e.g. in building and for insulation of remote-heating pipes and containers.

30 The following examples are designed to illustrate the invention without limiting its scope.

ExamplesExample 1 (Comparative example)

5 Formulation for rigid polyurethane foamed plastic

## Component A

50 parts by wt. O-tolylene diamine-started polyethers with propylene oxide

10 OH number = 400

50 parts by wt. of sugar-started polyethers with propylene oxide, OH number  
= 380

15 2 parts by wt. of H<sub>2</sub>O

20 2 parts by wt. of foam stabilizer B 8423 (made by Goldschmidt)

2 parts by wt. of Desmopac 726 b activator (made by Bayer AG)

20

## Component B

140 parts by wt. of crude MDI (NCO content: 31.5% by wt.)

25 100 parts by weight of component A were mixed with 12 parts by weight of cyclopentane and 140 parts by weight of component B in an agitator (1 000 rpm) at 20°C and compressed to 34 kg/m<sup>3</sup> in a closed mould.

Example 2 (according to the invention)

30

Formulation and processing as in Example 1, except that the blowing agent was a mixture of cyclopentane and n-butane in the molar ratio 85 : 15.

Example 3 (according to the invention)

As in Example 1 and 2.

5 The blowing-agent mixture consisted of cyclopentane and n-butane in the molar ratio 80 : 20.

Example 4 (according to the invention)

10 As in Examples 1 to 3.

Blowing-agent mixture of cyclopentane and i-butane in the molar ratio 90 : 10.

Results

15 The thermal conductivity and compressive strength of the foamed plastic slabs produced in Examples 1 to 3 was measured.

Table

Example	Thermal conductivity (mW/mK) to DIN 52 616, 24°C	Compressive strength (MPa) to DIN 53 421, under 10% compressive strain
1	21.1	0.14
2	21.3	0.17
3	21.5	0.19
4	21.4	0.18

As the results show, the compressive strength of rigid plastics having a given bulk density can be improved by admixtures of n-butane and i-butane.

Surprisingly there was no appreciable impairment of thermal conductivity. These blowing-agent mixtures can therefore be used for producing foamed plastics which largely retain their good insulating properties. They can be used preferably in refrigerator construction.

the first time in the history of the world, the *whole* of the human race, in all its parts, has been brought together in a single, common, and universal language.

Claims

1. Production of rigid polyurethane foamed plastics from polyols and polyisocyanates, blowing agents and optional foam auxiliary substances, 5 characterised in that the blowing agent is a mixture of 5 to 50 parts by weight of C<sub>3</sub> and/or C<sub>4</sub> alkanes and 50 to 95 parts by weight of cyclopentane.
2. Production of rigid polyurethane foamed plastics according to claim 1, 10 characterised in that the C<sub>4</sub>-alkane is n-butane.
3. Production of rigid polyurethane foamed plastics according to claim 1, 15 characterised in that the C<sub>4</sub>-alkane is isobutane.
4. Production of rigid polyurethane foamed plastics according to claims 1 to 3, 20 characterised in that in addition to the alkane mixture according to the invention, 0.5 to 4 parts by weight of water are used as the co-blowing agent.
5. Production of rigid polyurethane foamed plastics according to claim 4, 25 characterised in that preferably 1.5 to 3 parts by weight of water are used as the co-blowing agent.
6. Production of rigid polyurethane foamed plastics according to claims 1 to 5, characterised in that the polyol component is a mixture of 5 to 80 parts by weight of polyol based on aromatic amines.
7. Production of rigid polyurethane foamed plastics according to claim 6, 30 characterised by preferred use of 20 to 65 parts by weight of polyol based on aromatic amines.

8. A blowing-agent mixture for production of rigid polyurethane foamed plastic, characterised in that the mixture contains 5 to 50 parts by weight of C<sub>3</sub> and/or C<sub>4</sub>-alkanes and 50 to 95 parts by weight of cyclopentane.
- 5      9. A blowing-agent mixture according to claim 8, characterised in that the C<sub>3</sub> and/or C<sub>4</sub> alkanes therein are n-butane and/or isobutane.
- 10     10. Use of rigid polyurethane foamed plastics produced according to any of claims 1 to 9 as an intermediate layer for composite components and for foam filling of cavities in refrigerator construction.

## A method of producing rigid foamed plastics containing urethane groups

### Abstract

5 The description relates to production of rigid foamed plastics containing urethane groups, by use of a blowing-agent mixture containing 5 to 50 parts by weight of  $C_3$ , and/or  $C_4$  alkanes and 50 to 95 parts by weight of cyclopentane. There is also a description of use of the thus-produced rigid foamed plastics as composite components or for foam filling of cavities in refrigerator construction.

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**COMBINED DECLARATION AND POWER OF ATTORNEY**

**ATTORNEY DOCKET NO**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

## "PROCESS FOR PREPARING RIGID FOAMED MATERIALS CONTAINING URETHANE GROUPS"

the specification of which is attached hereto,

or was filed on December 02, 1996

as a PCT Application Serial No. PCT/EP96/05335

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

**195 46 461.3**      **Germany**      **December 13, 1995**  
(Number)      (Country)      (Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

(Application Serial No.) (Filing Date) (Status)  
(patented, pending, abandoned)

(Application Serial No.) (Filing Date) (Status)  
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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POST OFFICE ADDRESS		
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